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<p>(54) Title: ELECTRICAL DEVICE</p>			
<p>(57) Abstract</p> <p>An electrical device (1) containing a resistive element (9) composed of a conductive polymer composition which exhibits PTC behavior which is attached to two metal foil electrodes (11, 13). The device is prepared by a method which includes the steps of (a) cutting the device from a laminate containing the conductive polymer composition positioned between two metal foils; (b) exposing the device after the cutting step to at least one thermal excursion from a first temperature which is at most (T<sub>m</sub>-100) °C to a second temperature which is at most (T<sub>m</sub>-25) °C; and (c) crosslinking the conductive polymer composition after the thermal excursion. The devices of the invention have low resistance and are useful as circuit protection devices.</p>			

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## ELECTRICAL DEVICE

## BACKGROUND OF THE INVENTION

5 Field of the Invention

This invention relates to electrical devices comprising conductive polymers, and a method of making such devices.

10 Introduction to the Invention

Electrical devices which comprise conductive polymer compositions which exhibit PTC (positive temperature coefficient of resistance) behavior are well-known and are used as circuit protection devices and heaters. Such circuit protection devices, 15 containing low resistivity conductive polymer compositions, respond to changes in ambient temperature and/or current conditions. Under normal conditions, a circuit protection device remains in a low temperature, low resistance state when in series with a load in an electrical circuit. When exposed to an overcurrent or overtemperature condition, however, the device increases in resistance, effectively shutting down the 20 current flow to the load in the circuit. For many applications it is desirable that the device have as low a resistance as possible in order to minimize the contribution to the total resistance of the electrical circuit during normal operation. Furthermore, a low resistance allows the device to have a higher hold current, i.e. the largest steady state current that, under specified ambient conditions, can be passed through a circuit protection device 25 without causing the device to "trip" into the high resistance state. Although low resistance devices can be made from a given composition by changing dimensions, e.g. making the distance between the electrodes very small or the device area very large, small devices are preferred. Such devices occupy less space on a circuit board and generally have desirable thermal properties.

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## BRIEF SUMMARY OF THE INVENTION

The most common technique to achieve a small device is to use a composition that has a low resistivity. It is known that various processing steps, e.g. irradiation, 35 encapsulation, or exposure to elevated temperature during solder reflow, can increase the resistivity of the composition during device fabrication. Therefore, it is desirable to use

processing techniques which increase the resistivity of the composition as little as possible so that the final device will have a low resistance. We have now found a particular sequence of processing steps which helps in producing a device with a low resistance. In a first aspect this invention provides an electrical device which comprises

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- (A) a resistive element which is composed of a conductive polymer composition which exhibits PTC behavior and which comprises
  - (1) a polymeric component having a crystallinity of at least 20% and a melting point  $T_m$ , and
  - (2) dispersed in the polymeric component, a particulate conductive filler; and
- 10 (B) two electrodes which (i) are attached to the resistive element, (ii) comprise metal foils, and (iii) can be connected to a source of electrical power,

the device having been prepared by a method which comprises the steps of

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- (a) cutting a device from a laminate comprising the conductive polymer composition positioned between two metal foils;
- (b) exposing the device after the cutting step to at least one thermal excursion from a first temperature which is at most  $(T_m - 100)^\circ\text{C}$  to a second temperature which is at most  $(T_m - 25)^\circ\text{C}$ ; and
- 25 (c) crosslinking the conductive polymer composition after the thermal excursion.

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In a second aspect, the invention provides a method of making an electrical device which comprises

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- (A) a resistive element which (i) has a thickness of at most 0.51 mm, (ii) is crosslinked to the equivalent of at least 2 Mrads, and (iii) is composed of a conductive polymer composition which comprises

(1) a polymeric component having a crystallinity of at least 20% and a melting point  $T_m$ , and

5 (2) dispersed in the polymeric component, a particulate conductive filler; and

(B) two electrodes which (i) are attached to the resistive element, (ii) comprise metal foils, and (iii) can be connected to a source of electrical power,

10 said method comprising

(a) cutting a device from a laminate comprising the conductive polymer composition positioned between two metal foils,

15 (b) exposing the device after the cutting step to at least one thermal cycle from a first temperature which is at most  $(T_m - 100)^\circ\text{C}$  to a second temperature which is at most  $(T_m - 25)^\circ\text{C}$  and back to the first temperature, and

20 (c) crosslinking the conductive polymer composition after the thermal cycle step.

In third aspect, the invention provides a battery assembly which comprises

25 (I) a battery which comprises first and second terminals; and

(II) a device according to the first aspect of the invention which further comprises

30 (C) a first conductive lead which is attached to the first electrode, and

(D) a second conductive lead which is attached to the second electrode;

35 said device having been placed in contact with the first terminal of the battery so that the first conductive lead is in physical and electrical contact with that terminal.

## BRIEF DESCRIPTION OF THE DRAWINGS

The invention is illustrated by the drawings in which Figure 1 is a cross-sectional view of a device of the invention;

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Figure 2 is a top view of a device of the invention;

Figure 3 is a top view of a first conductive lead which forms part of a device of the invention;

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Figure 4 is a top view of a device of the invention with a second conductive lead attached;

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Figure 5 is a top view of a device of the invention comprising an insulating layer; and

Figure 6 is a graph showing the resistance distribution for devices made by a method of the invention and two comparative methods.

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## DETAILED DESCRIPTION OF THE INVENTION

The electrical device of the invention comprises a resistive element composed of a conductive polymer composition, and first and second metal foil electrodes which are attached to, and sandwich, the resistive element. The conductive polymer composition comprises a polymeric component, and dispersed therein, a particulate conductive filler. The polymeric component comprises one or more polymers, one of which is preferably a crystalline polymer having a crystallinity of at least 20% as measured in its unfilled state by a differential scanning calorimeter. Suitable crystalline polymers include polymers of one or more olefins, particularly polyethylene such as high density polyethylene; 25 copolymers of at least one olefin and at least one monomer copolymerisable therewith such as ethylene/acrylic acid, ethylene/ethyl acrylate, ethylene/vinyl acetate, and ethylene/butyl acrylate copolymers; melt-shapeable fluoropolymers such as polyvinylidene fluoride (PVDF) and ethylene/tetrafluoroethylene copolymers (ETFE, including terpolymers); and blends of two or more such polymers. For some applications 30 it may be desirable to blend one crystalline polymer with another polymer, e.g. an elastomer or an amorphous thermoplastic polymer, in order to achieve specific physical or 35

thermal properties, e.g. flexibility or maximum exposure temperature. The polymeric component generally comprises 40 to 90% by volume, preferably 45 to 80% by volume, especially 50 to 75% by volume of the total volume of the composition.

5        The particulate conductive filler which is dispersed in the polymeric component may be any suitable material, including carbon black, graphite, metal, metal oxide, conductive coated glass or ceramic beads, particulate conductive polymer, or a combination of these. The filler may be in the form of powder, beads, flakes, fibers, or any other suitable shape. The quantity of conductive filler needed is based on the  
10      required resistivity of the composition and the resistivity of the conductive filler itself. For many compositions the conductive filler comprises 10 to 60% by volume, preferably 20 to 55% by volume, and for low resistivity compositions used for low resistance circuit protection devices, especially 25 to 50% by volume of the total volume of the composition.

15       The conductive polymer composition may comprise additional components, such as antioxidants, inert fillers, nonconductive fillers, radiation crosslinking agents (often referred to as prorads or crosslinking enhancers, e.g. triallyl isocyanurate), stabilizers, dispersing agents, coupling agents, acid scavengers (e.g.  $\text{CaCO}_3$ ), or other components.

20       These components generally comprise at most 20% by volume of the total composition. Dispersion of the conductive filler and other components into the polymeric component may be achieved by any suitable means of mixing, e.g. melt-processing or solvent-mixing. The mixed composition can be melt-shaped by any suitable method, e.g. melt-extrusion, injection-molding, compression-molding, or sintering, in order to produce a  
25      resistive element. The element is preferably laminar and may be of any shape, e.g. rectangular, square, circular, or annular. The resistive element often has a thickness of at most 1.02 mm (0.040 inch), and, for many applications, is much thinner, i.e. has a thickness of at most 0.51 mm (0.020 inch), preferably at most 0.38 mm (0.015 inch).

30       The composition used in the resistive element exhibits positive temperature coefficient (PTC) behavior, i.e. it shows a sharp increase in resistivity with temperature over a relatively small temperature range. In this application, the term "PTC" is used to mean a composition or device which has an  $R_{14}$  value of at least 2.5 and/or an  $R_{100}$  value of at least 10, and it is preferred that the composition or device should have an  $R_{30}$  value of at least 6, where  $R_{14}$  is the ratio of the resistivities at the end and the beginning of a  $14^\circ\text{C}$  range,  $R_{100}$  is the ratio of the resistivities at the end and the beginning of a

100°C range, and R<sub>30</sub> is the ratio of the resistivities at the end and the beginning of a 30°C range. Generally the compositions used in devices of the invention show increases in resistivity which are much greater than those minimum values.

5        Suitable conductive polymer compositions are disclosed in U.S. Patent Nos. 4,237,441 (van Konynenburg et al), 4,545,926 (Fouts et al), 4,724,417 (Au et al), 4,774,024 (Deep et al), 4,935,156 (van Konynenburg et al), 5,049,850 (Evans et al), 5,250,228 (Baigrie et al), 5,378,407 (Chandler et al), 5,451,919 (Chu et al), 5,582,770 (Chu et al), 5,747,147 (Wartenberg et al), and 5,801,612 (Chandler et al), and in  
10      commonly assigned International Publication No. WO96/29711 (Raychem Corporation, published September 26, 1997).

15      The resistive element is attached to first and second laminar electrodes which are attached to the first face and the second face, respectively, of the resistive element. It is preferred that the conductive polymer composition be extruded or otherwise formed into a sheet onto which the electrodes may be attached to form a laminate, i.e. the conductive polymer is sandwiched between the foils. Both first and second electrodes comprise a conductive material, and are preferably metal in the form of a foil, e.g. nickel, copper, brass, stainless steel, or an alloy of one or more of these metals, although one or both of  
20      the electrodes may comprise a conductive paint or graphite layer. A tie layer, e.g. a conductive adhesive, may be used to attach the electrode to the resistive element. It is particularly preferred that the first and second electrodes comprise an electrodeposited metal foil, e.g. nickel, copper, or nickel-coated copper. Appropriate electrodes are disclosed in U.S. Patents Nos. 4,689,475 (Matthiesen) and 4,800,253 (Kleiner et al), and  
25      in International Publication No. WO95/34081 (Raychem Corporation, published December 14, 1995).

30      The device may also comprise an insulating layer, which provides electrical and environmental protection to the device. The insulating layer generally covers some or all of the metal foil electrodes and any exposed surfaces of the resistive element. Suitable insulating materials include polymers such as polyamide, polybutylene terephthalate, a polyester, polyethylene, polyvinylidene fluoride, liquid crystalline polymers, or an epoxy resin.

35      In the method of the invention, in a cutting step, the device is cut from a laminate comprising the conductive polymer composition positioned between two metal foils. In

this application, the term "cutting" is used to include any method of isolating or separating the resistive element of the device from the laminate, e.g. dicing, punching, shearing, cutting, etching and/or breaking as described in International Publication No. WO95/34084 (published December 14, 1995).

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The device is then subjected to a thermal treatment step. The thermal treatment step comprises at least one excursion from a first temperature  $T_1$  to a second temperature  $T_2$ . Preferably the thermal treatment step includes a return to the first temperature after exposure to the second temperature, thus creating a cycle from  $T_1$  to  $T_2$  to  $T_1$ . The first 10 temperature is at most  $(T_m - 100)^\circ\text{C}$ , preferably at most  $(T_m - 120)^\circ\text{C}$ , particularly at most  $(T_m - 150)^\circ\text{C}$ , where  $T_m$  is the melting point of the polymeric component, as measured by the peak of the endotherm of a differential scanning calorimeter. When there is more than one peak, for example when the polymeric component comprises a mixture of crystalline polymers,  $T_m$  is defined as the temperature of the highest temperature peak. The second 15 temperature is at most  $(T_m - 25)^\circ\text{C}$ , preferably at most  $(T_m - 35)^\circ\text{C}$ , particularly at most  $(T_m - 50)^\circ\text{C}$ . It is important that the first temperature be at a temperature above the glass transition temperature  $T_g$  of the polymeric component.  $T_1$  is often at a temperature below room temperature, i.e. less than  $20^\circ\text{C}$ . In a preferred embodiment of the method, the device is exposed to at least two thermal cycles, preferably at least three thermal cycles. 20 For some applications, the device may be exposed to many more thermal cycles, e.g. six thermal cycles. During the thermal treatment step, for the thermal excursion or for each thermal cycle, the device is held at both the first and the second temperatures for sufficient time to ensure that the entire device reaches the designated temperature. The time period during which the device is held may be the same or different at  $T_1$  and  $T_2$ , but 25 is generally at least 1 minute, preferably at least 3 minutes, more preferably at least 5 minutes, particularly at least 10 minutes, more particularly at least 15 minutes, especially at least 30 minutes, e.g. 60 minutes, as measured from the time the device reaches the designated temperature. Any suitable heat source may be used during the thermal treatment step, e.g. an oven (particularly a programmable oven) or other environmental 30 chamber, or a heat lamp. The rate of temperature increase from  $T_1$  to  $T_2$  (and back to  $T_1$ , if present), may be any convenient rate, e.g. 2 to  $30^\circ\text{C}/\text{minute}$ . When the thermal treatment step is a thermal cycle, the rate from  $T_1$  to  $T_2$  may be the same as or different from the rate from  $T_2$  to  $T_1$ .

Following the thermal treatment step, the conductive polymer composition is crosslinked. Crosslinking can be accomplished by chemical means or by irradiation, e.g. using an electron beam or a  $\text{Co}^{60} \gamma$  irradiation source. The level of crosslinking depends on the required application for the composition, but is generally less than the equivalent 5 of 200 Mrads, and is preferably substantially less, i.e. from 1 to 20 Mrads, preferably from 1 to 15 Mrads, particularly from 2 to 10 Mrads for low voltage (i.e. less than 60 volts) applications. Useful circuit protection devices for applications of less than 30 volts can be made by irradiating the device to at least 2 Mrads but at most 10 Mrads.

10 For many applications, it is necessary to attach at least one conductive lead, i.e. a first conductive lead, to one of the metal foil electrodes. Often first and second conductive leads are attached to first and second electrodes, respectively. The conductive leads allow easy connection of the electrodes to the source of electrical power, e.g. a battery or a power source, or into the circuit, and can be used to control the thermal output 15 of the device. The conductive leads, which often are supplied as part of a lead frame for ease in manufacture, are preferably attached to the electrodes by means of an intermediate layer, e.g. solder or a conductive adhesive. This lead attachment step preferably occurs after the cutting step and before the thermal treatment step. Other assembly processes, e.g. application of the electrically insulating layer such as an epoxy resin or other 20 polymer, are preferably conducted during an assembly step, which includes the lead attachment step and occurs after the cutting step and before the thermal treatment step.

Following the method of the invention, devices in which the conductive polymer has a low resistivity, i.e. less than 100 ohm-cm, preferably less than 20 ohm-cm, 25 particularly less than 10 ohm-cm, more particularly less than 5 ohm-cm, especially less than 2 ohm-cm, e.g. less than 1 ohm-cm, can be prepared. For most applications, the device generally has a resistance at 20°C of less than 1 ohm, preferably less than 0.5 ohms, particularly less than 0.25 ohms, e.g. 0.050 to 0.150 ohms.

30 Devices of the invention are particularly suitable for use in a battery assembly, in which the device is preferably attached to the first or second terminal of a battery. The attachment may be directly between the battery and the first or second electrode, or between the battery and the first conductive lead which is attached to the first electrode, or the second conductive lead which is attached to the second electrode. For some 35 batteries, in which either the first and/or the second terminal is in the form of a "button", the device is physically and electrically attached to a button terminal. The device may be

attached to either the negative or the positive terminal of the battery. Batteries suitable for use include nickel-cadmium, nickel-metal hydride, alkaline, or lithium batteries. Often the battery assembly comprises two or more batteries. Such a battery assembly is shown in International Publications Nos. WO97/06538 (K.K. Raychem, published 5 February 20, 1997) and WO98/20567 (Raychem Corporation, published May 14, 1998).

The invention is illustrated by the drawings in which Figure 1 shows in cross-section electrical device 1 of the invention comprising PTC component 3, first conductive lead 15, second conductive lead 18, and insulating material 23. PTC component 3 10 comprises first electrode 11, second electrode 13 and resistive element 9 composed of a conductive polymer sandwiched therebetween. In PTC component 3 shown in Figure 1, both electrodes opposing each other form first surface 43 and second surface 41.

A top view of PTC component 3 is shown in Figure 2. PTC component 3 15 comprises surfaces 41 and 43, outer periphery 5 and inner periphery 7 and is in the form of a disk with an opening 27 in the center. The inner periphery 7 defines the opening 27.

Figure 3 shows a top view of first conductive lead 15, which has a first part 16 which is attached to first electrode 11, and a second part 17 which extends across opening 20 27 of PTC component 3. In this embodiment, first part 16 covers the whole surface of first electrode 11. Second part 17 of first lead 15, which covers at least a part of opening 27, is used to make direct electrical contact, via solder, pressure, or a weld, to a button terminal of a battery. The second part 17 covers at least a part of opening 27.

25 Figure 4 shows a top view of second conductive lead 18, which comprises first part 19 and second part 21. First part 19 is attached to second electrode 13 and covers at least part of the surface of second electrode 13. Second part 21 extends away from outer periphery 5 and can be bent, if necessary, to make electrical contact to a second battery or other electrical component. First and second conductive leads 15,18 can be made from 30 any suitable material, e.g. nickel, stainless steel, copper, or an alloy such as brass or bronze. For ease of manufacture second lead 18 is often part of a lead frame.

Figure 5 shows a top view of device 1 which has been encapsulated by electrically insulating layer 23. When a positive terminal of a battery is placed inside opening 27', no 35 short circuit occurs because no electrical contact is made between the terminal and PTC component 3.

The invention is illustrated by the following examples, in which Examples 1 and 2 are comparative examples. The following steps were conducted for each example:

5    Preparation of PTC Device

A first compound was prepared by preblending 54% by weight carbon black (Raven<sup>TM</sup> 430U, available from Columbian Chemicals) with 46% by weight high density polyethylene (Petrothene<sup>TM</sup> LB832, available from Millenium; "HDPE") in a Henschel 10 blender, mixing the blend in a Buss kneader, and extruding into pellets. A second compound was prepared by preblending 51.4% by weight carbon black with 48.6% by weight HDPE in the same way. Pellets of the first and second compounds were preblended to give a final compound having 52.7% by weight carbon black and 47.3% by weight HDPE, and this compound was extruded through a sheet die using an Egan 15 extruder to give a sheet with a thickness of 0.25 mm (0.010 inch). The extruded sheet was laminated between two layers of electrodeposited nickel foil having a thickness of about 0.033 mm (0.0013 inch) (available from Fukuda) by using a press set at 200°C. The laminated sheet was solder-dipped into a solder formulation of 63% lead/37% tin 20 heated to 230°C, and devices having the shape shown in Figure 2 were punched from the laminated sheet.

Assembly of Device

First and second conductive leads, as shown in Figures 3 and 4, were attached to 25 the PTC device by means of 63/37 lead/tin solder, which was reflowed in a hot-air oven which increased in temperature from 30°C to a maximum temperature of 230°C over about 2 minutes. An insulating layer of liquid crystalline polymer was then applied by means of transfer or injection molding.

30    Irradiation

Devices were irradiated by using a cobalt 60  $\gamma$  irradiation source to give a total irradiation of 14 Mrad.

Temperature Cycling

The devices were thermally cycled six times, each cycle being from -40 to 80 to -40°C at a rate of 1°C/minute with a 60 minute dwell at -40°C and 80°C.

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Examples 1 and 2 (Comparative)

Device preparation was conducted as shown in Table I, with numbers 1 to 4 indicating the sequential order of the process steps as they were conducted. The 10 resistance at 25°C was measured for one hundred devices prepared for each example. The resulting devices had an average resistance at least 5% higher than that of devices of the invention (Example 3).

Example 3

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Device preparation was conducted as shown in Table I. The resulting devices had a resistance lower than that of conventional devices. A comparison of the resistance distribution of the devices is shown in Figure 6.

<u>Example</u>	<u>Device Preparation Steps</u>				<u>Resistance</u> (mohms)
	<u>Preparation</u>	<u>Assembly</u>	<u>Irradiation</u>	<u>Temp. Cycle</u>	
1	1	2	3	4	53.3
2	1	3	2	4	64.9
3	1	2	4	3	49.0

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Although the various figures and descriptions in this specification relate to specific embodiments of the invention, it is to be understood that where a specific feature is disclosed in the context of a particular figure, such feature can also be used, to the 25 extent appropriate, in the context of another figure, in combination with another feature, or in the invention in general. It will be understood that the above-described arrangements of apparatus and the methods therefrom are merely illustrative of applications of the principles of this invention and many other embodiments and modifications may be made without departing from the spirit and scope of the invention 30 as defined in the claims.

What is claimed is:

1. An electrical device which comprises
  - (A) a resistive element which is composed of a conductive polymer composition which exhibits PTC behavior and which comprises
    - (1) a polymeric component having a crystallinity of at least 20% and a melting point  $T_m$ , and
    - (2) dispersed in the polymeric component, a particulate conductive filler; and
  - (B) two electrodes which (i) are attached to the resistive element, (ii) comprise metal foils, and (iii) can be connected to a source of electrical power,

the device having been prepared by a method which comprises the steps of

  - (a) cutting a device from a laminate comprising the conductive polymer composition positioned between two metal foils;
  - (b) exposing the device after the cutting step to at least one thermal excursion from a first temperature which is at most  $(T_m - 100)^\circ\text{C}$  to a second temperature which is at most  $(T_m - 25)^\circ\text{C}$ ; and
  - (c) crosslinking the conductive polymer composition after the thermal excursion.
  2. A device according to claim 1 wherein the method further comprises attaching at least one conductive lead to one of the metal foil electrodes after step (a) and before step (b).
  3. A device according to claim 1 or 2 which has a resistance of at most 0.100 ohm.
  4. A device according to claim 1, 2, or 3 which has been crosslinked in step (c) to the equivalent of 1 to 20 Mrads.

5. A device according to any one of the preceding claims wherein the polymeric component comprises polyethylene, an ethylene copolymer, or a fluoropolymer.
6. A device according to any one of the preceding claims wherein in step (b) the device is exposed to at least one thermal cycle from the first temperature to the second temperature and back to the first temperature.
7. A device according to any one of the preceding claims wherein in step (b) the first temperature  $T_1$  is less than 23°C.
8. A method of making an electrical device according to claim 1 wherein
  - (1) the resistive element (i) has a thickness of at most 0.51 mm and (ii) is crosslinked to the equivalent of at least 2 Mrads, and
  - (2) in step (b), the device is exposed to at least one thermal cycle from a first temperature which is at most  $(T_m - 100)^\circ\text{C}$  to a second temperature which is at most  $(T_m - 25)^\circ\text{C}$  and back to the first temperature after the cutting step.
9. A method according to claim 8 wherein in step (b) the device is exposed to at least three thermal cycles.
10. A battery assembly which comprises
  - (1) a battery which comprises first and second terminals; and
  - (2) a device according to claim 1 which further comprises (a) a first conductive lead which is attached to the first electrode, and (b) a second conductive lead which is attached to the second electrode;

said device having been placed in contact with the first terminal of the battery so that the first conductive lead is in physical and electrical contact with that terminal.

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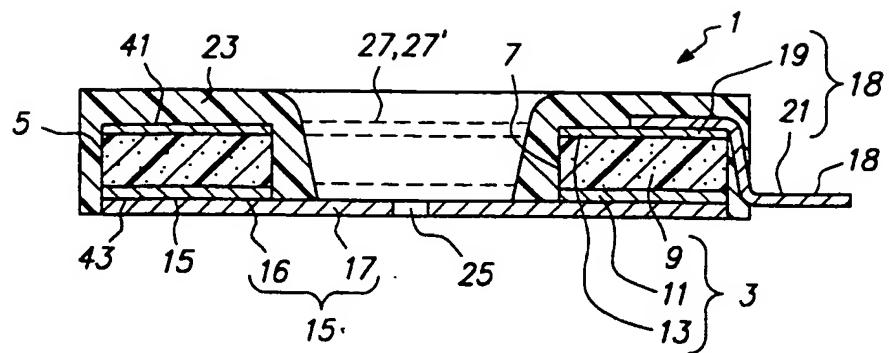


FIG. 1

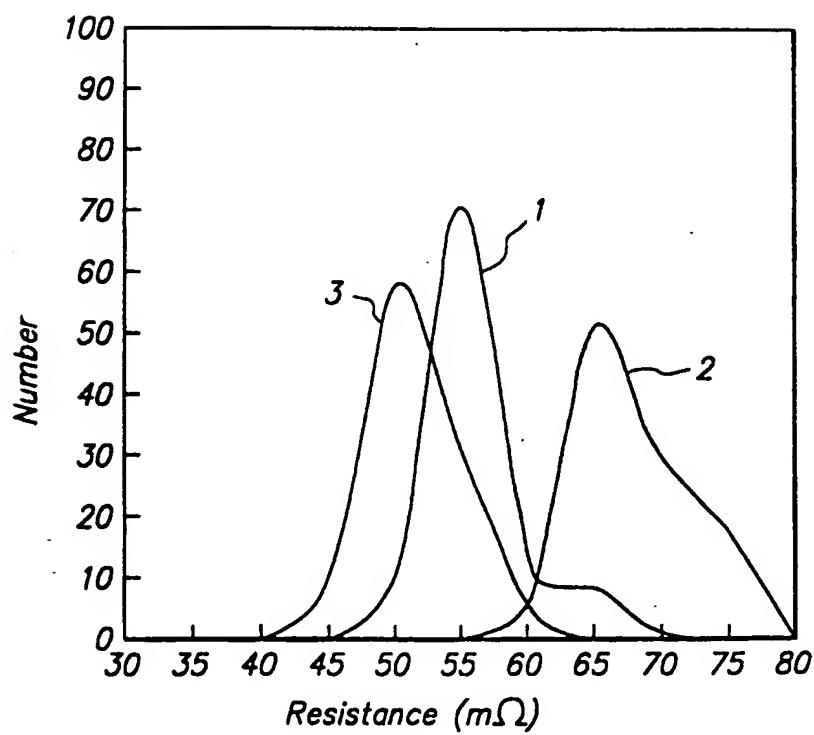
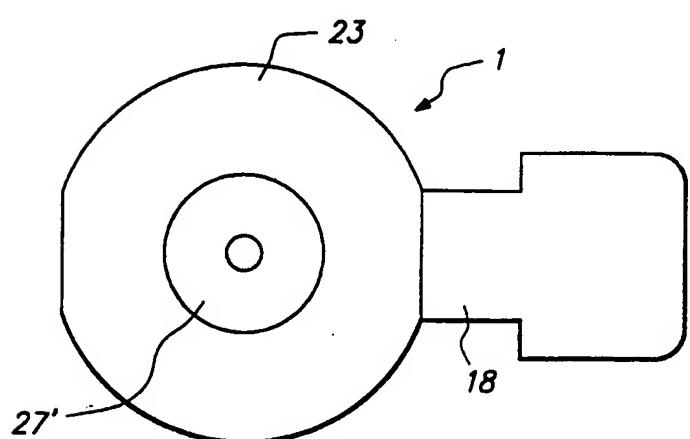
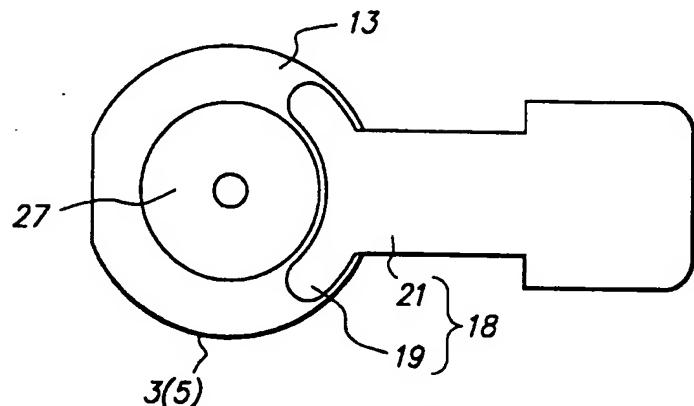
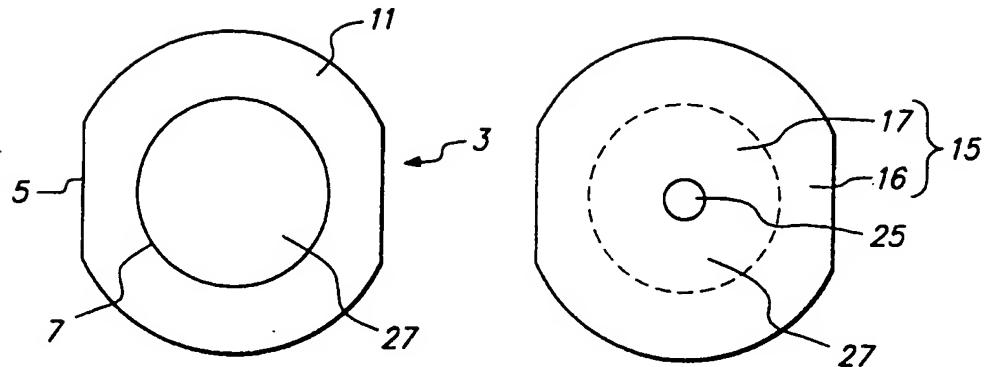


FIG. 6

2/2



# INTERNATIONAL SEARCH REPORT

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**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 6 H01C7/02 H01C17/30

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 H01C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 522 863 A (DAITO TSUSHINKI KK) 13 January 1993 see page 7, line 35 - page 7, line 44; claims 4,6,8,10; table 5	1,3,5-8
A	---	4
Y	EP 0 730 282 A (UNITIKA LTD) 4 September 1996 see the whole document	1,3,5-8
A	---	9
P,A	EP 0 844 622 A (RAYCHEM KK) 27 May 1998 cited in the application see page 6, line 36 - page 9, line 40; claims 1-8; figures 1-10	1,3,5,9
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	-/-	

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

\* Special categories of cited documents :

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Date of the actual completion of the international search

9 March 1999

Date of mailing of the international search report

16/03/1999

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## INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/26328

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 484 138 A (DAITO TSUSHINKI KK) 6 May 1992 see the whole document -----	1,5
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Information on patent family members

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